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Half-lives of isomeric states in Pb^{202}, Pb^{203}, Pb^{206} and Tl^{202}

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With 7 figures in the text

SUMMARY

The isomeric transitions in Pb^{202} ($E4$, $E5$), Pb^{203} ($M4$), Pb^{206} ($E3$) and Tl^{202} ($E3$) are associated with high multipolarities. The half-lives of Pb^{202m} and Pb^{203m} were accurately measured by means of an automatic recording arrangement. In the case of Pb^{206m} and Tl^{202m} a modified delayed-coincidence technique was used. The following half-lives were found:

Pb^{202m} : 3.62 ± 0.03 hours

Pb^{203m} : 6.09 ± 0.10 sec

Pb^{206m} : 128 ± 5 μsec

Tl^{202m} : 600 ± 200 μsec

Introduction

The half-lives measured in this investigation are all associated with transitions of high multipole order.

The $9-$ state in Pb^{202} decays by $E5$ and $E4$ transitions, Fig. 1A. This decay has been extensively studied by McDonell *et al.* [1]. Their high resolution measurements were always extended over several half-lives ($T_{\frac{1}{2}} = 3.7$ h). The accuracy in their results is therefore to a high degree depending on the error in the half-life determination. An accuracy of at least 1% seemed necessary.

It has been shown by Stockendal *et al.* [2] and by Andersson *et al.* [3] that the radial matrix elements of the $M4$ transitions in Pb^{197}, Pb^{199}, Pb^{201}, Pb^{203} and Pb^{207} show a remarkable constancy. The matrix element of Pb^{203}, however, was found to be somewhat larger than the others. Since the half-life of Pb^{203m} is rather short and was not very accurately known ($6,5 \pm 0.5$ sec), it might be possible that the deviation of the matrix element in this case was entirely due to experimental errors in the half-life determination. In order to settle this question we have therefore remeasured this half-life with higher accuracy.

In the decay of Pb^{202m} [1] there is a 460 keV $E3$ γ-ray converted in Tl. That transition should be associated with a half-life in the 100 μsec region. When bombarding natural Tl with 20 MeV photons, Vegors and Axel [4] found two isomeric states of half-lives 530 ± 50 μsec and 65 ± 5 μsec. Since natural Tl consists of 30% Tl^{203} and 70% Tl^{205} and (γ,n) processes are the most probable ones for 20 MeV

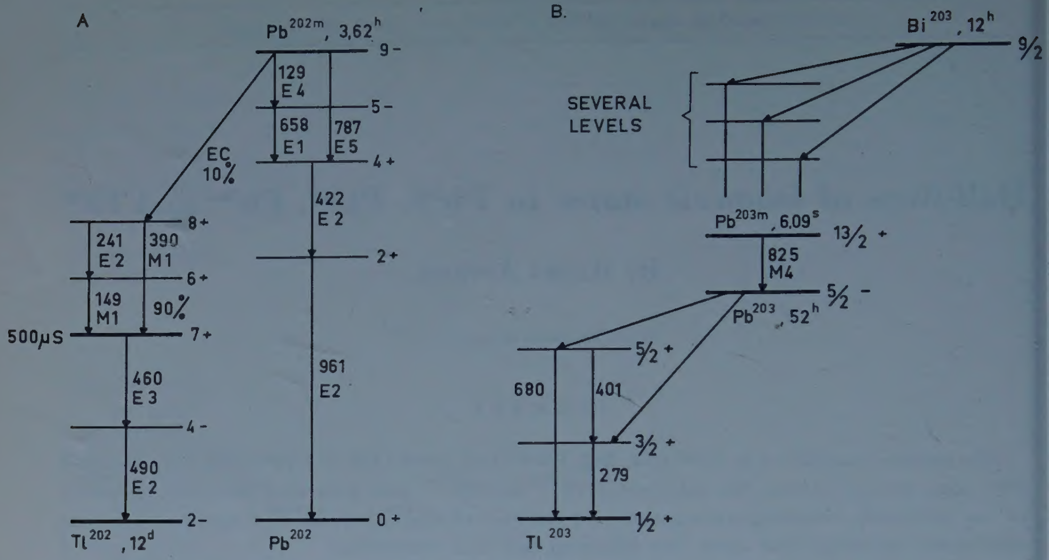

Fig. 1. Simplified decay schemes of Pb^{202}, Pb^{203} and Tl^{202}, showing levels and transitions essential for this work.

photons, it seemed very likely that these isomeric states belonged to Tl^{202} and Tl^{204}. By measuring delayed coincidences in a Pb^{202m} sample we hoped to be able to observe one of the activities found by Vegors and Axel. Owing to the large difference in half-lives no high accuracy was needed to determine which of the two half-lives is associated with Tl^{202}.

In order to test the delayed-coincidence equipment utilized in the above measurement we used a Bi^{206} sample ($T_{\frac{1}{2}} = 6.4$ d). In the radiation from this sample one can study the half-life of the 7 − state in Pb^{206} which decays by the emission of two $E3$ gamma rays [5]. It has been pointed out by Alburger and Pryce [6] that when measuring the half-life of the 7 − state by delayed coincidence measurements, the coincidence efficiency is very high, because several prompt gamma rays are emitted before and after this level. The half-life found for this state indicates the accuracy that can be obtained with the simple apparatus used.

Measurements and results

Pb^{202m}

The principal levels and transitions in the decay of Pb^{202m} [1] are shown in Fig. 1 A. The half-life was given by Wapstra [7] to be 3.7 h.

The samples used for the half-life measurements were prepared by bombarding lead with protons in the Uppsala cyclotron and also by bombarding thallium with deutrons in the Stockholm cyclotron. In the first case bismuth is formed, which decays into lead by electron capture. Several lead isotopes are formed and the measurement to be described here was performed with a sample in which Pb^{202} had been enriched by electromagnetic isotope separation. As can be seen from the successive γ-spectra of Fig. 2, a certain amount of Pb^{203} is present. This was fortunate because

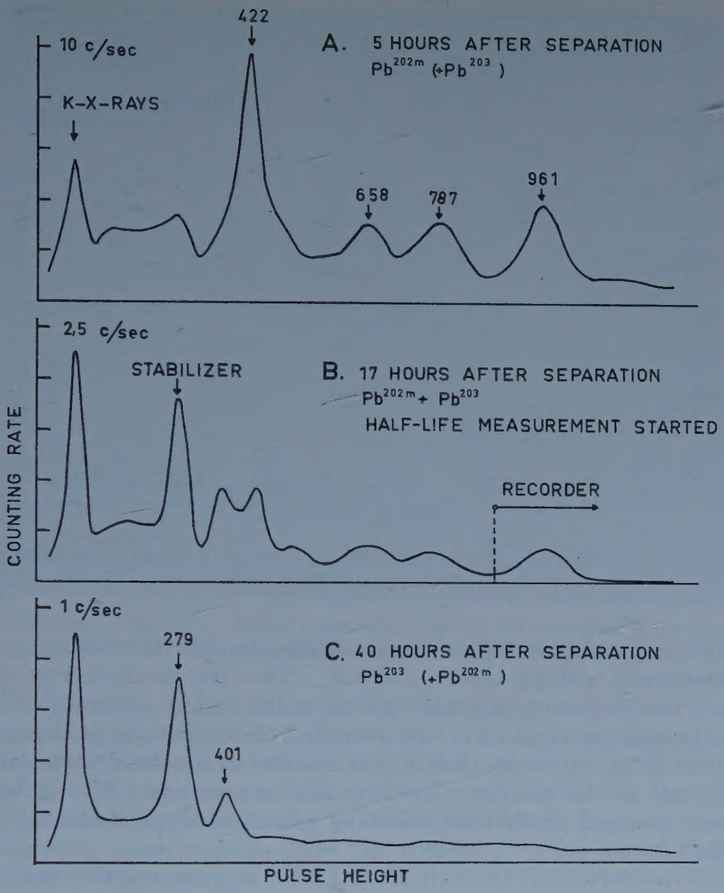

Fig. 2. Scintillation spectra recorded at the same time as the decay of Pb^{202m} was measured with an identical sample. The channel width was 2 %.

the longer-lived 279 keV γ-ray (Fig. 2C) from Pb^{203} ($T_{\frac{1}{2}} = 52$ h) could be used as a reference line during the experiment. No other γ-rays with an energy higher than 850 keV were present in detectable amounts, excepting the 961 keV-line in Pb^{202}.

The decay of this line was followed by means of a single-channel scintillation spectrometer [8] adjusted to accept all pulses corresponding to energies higher than 850 keV (Fig. 2B). The pulses were counted with a scale-of-800. An Esterline–Angus recorder was connected parallel to the mechanical register and thus marked the arrival of each 800th pulse on the continuously running chart of the recorder. The recorder also received time markers of opposite polarity every 10 minutes. In this way the decay was continuously followed and recorded for 44 hours, corresponding to 12 half-lives. The last part of the time gave a good determination of the background. To eliminate drift in the spectrometer a stabiliser of the de Waard type [9] was used and accepted the 279 keV-line from Pb^{203} (Fig. 2B).

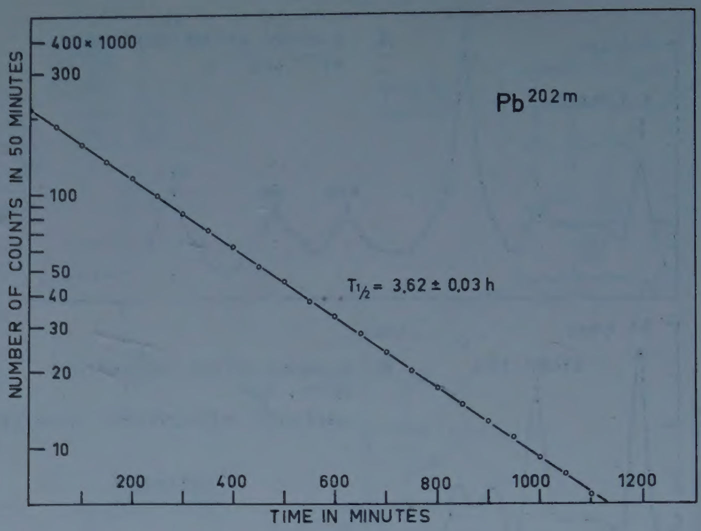

Fig. 3. Decay of Pb^{202m}. The background has been subtracted. The statistical uncertainty is less than the diameter of the points.

The background was about 5% of the counting-rate when the measurement started and was determined with an accuracy of 0.3%. The time markers were obtained by scaling down the frequency of a tuning fork which had an accuracy of 0.1%. Two measurements were made and the data recorded on the Esterline–Angus chart were converted into decay curves by plotting the number of counts in successive intervals (Fig. 3), as well as the number of counts left, versus time. Five different decay curves were drawn and yielded the following values for the half-life: 3.58, 3.63, 3.62, 3.63, and 3.64 hours.

From these measurements the half-life of Pb^{202m} is given as 3.62 ± 0.03 hours. The limits of error have been set to include small systematic errors not known at present.

Pb^{203m}

In an experiment where the Pb activity was milked from an ion exchange column loaded with decaying Bi isotopes, Stockendal *et al.* [2] found the half-life of Pb^{203m} to be 6.5 ± 0.5 sec. The same method was used in our experiment and the counts were recorded in the same way (Fig. 4) as has been described for the Pb^{202} measurements. The stabilizer was not used because of the short measuring time. The discriminator channel was adjusted to accept γ-rays of 825 keV energy, which is the only γ-energy found in the decay of Pb^{203m}, Fig. 1B. Time markers were recorded every two seconds.

The Pb^{203m} activity brought from the column to the detector decayed completely in about 150 seconds, and by extending the measurement over 300 seconds or more the background was accurately determined. The maximum source strength that can be used in a measurement of this type is determined by the time resolution of the discriminator (about 2 μsec). The total number of disintegrations that can be counted in each experiment is therefore limited. This fact, in combination with the way of recording (writing down the actual time for each 800th count), makes it most

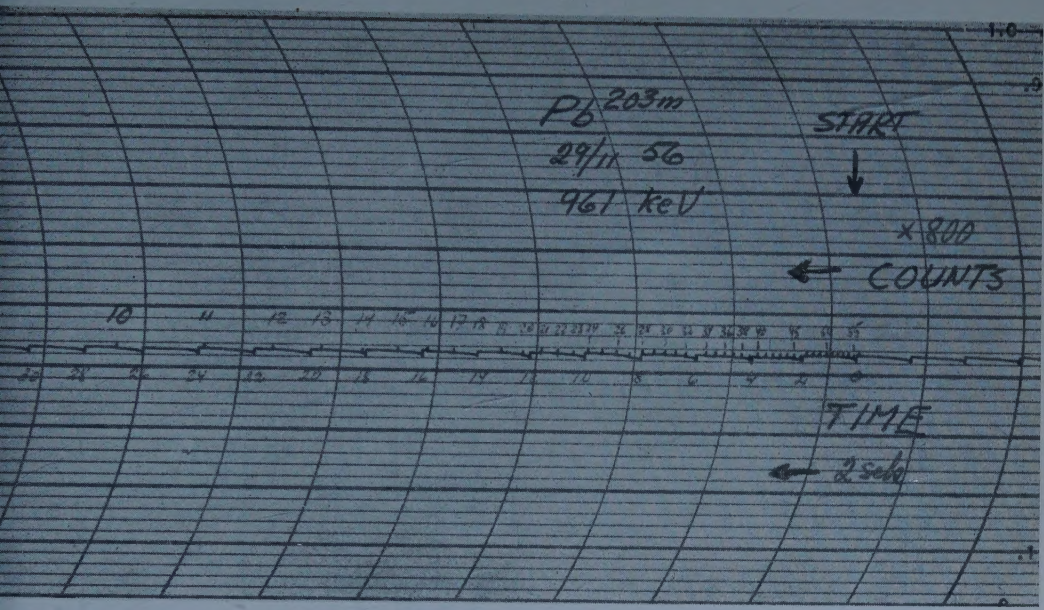

Fig. 4. Reproduction of Esterline–Angus recording, showing the first part of the decay of Pb^{203m}. Erratum: The spectrometer was accepting B25 keV γ-rays.

convenient to plot the decay curve as the "number of counts left", versus time. If a plot of this type is made in a linear scale, a constant background will give a sloping straight line that can be extrapolated back to zero time. After the background found from that plot has been subtracted, the decay curve is a straight line in a logarithmic plot and the half-life can be determined in the ordinary way, Fig. 5.

Four successive measurements gave the following values of the half-life: 6.08, 6.10, 6,12, and 6.06 seconds.

Systematic errors from the source geometry and changes in photomultiplier gain due to fatigue effects must be taken into account. The drops from the column were caught in a glass tube and brought to the detector. It is possible that some liquid on the walls of the tube changed its position during the measurement. Certain photomultipliers change their gain with the counting rate [10]. Unfortunately it was not investigated whether this was the case with the tube used in these measurements. However, if a change in gain had shifted the peak position relative to the discriminator channel-setting, it seems likely that the measured decay curve should have deviated from a straight line; but no such effect was observed.

Taking possible systematic errors into account, the half-life of Pb^{203m} is given as 6.09 ± 0.10 seconds. This means that the radial matrix element of the $M4$ transition in Pb^{203} is 4.3 ± 0.1 and that in fact this matrix element is somewhat larger than for Pb^{197}, Pb^{199}, Pb^{201} and Pb^{207}, for which the matrix elements are 3.8 ± 0.5, 3.7 ± 0.2, 3.5 ± 0.2, and 3.6 ± 0.1 respectively [2, 3].

Pb^{206m} and Tl^{202m}

As has already been mentioned in the introduction the purpose of this measurement was to determine whether the 7+ state in Tl^{202} (Fig. 1A) has a half-life of

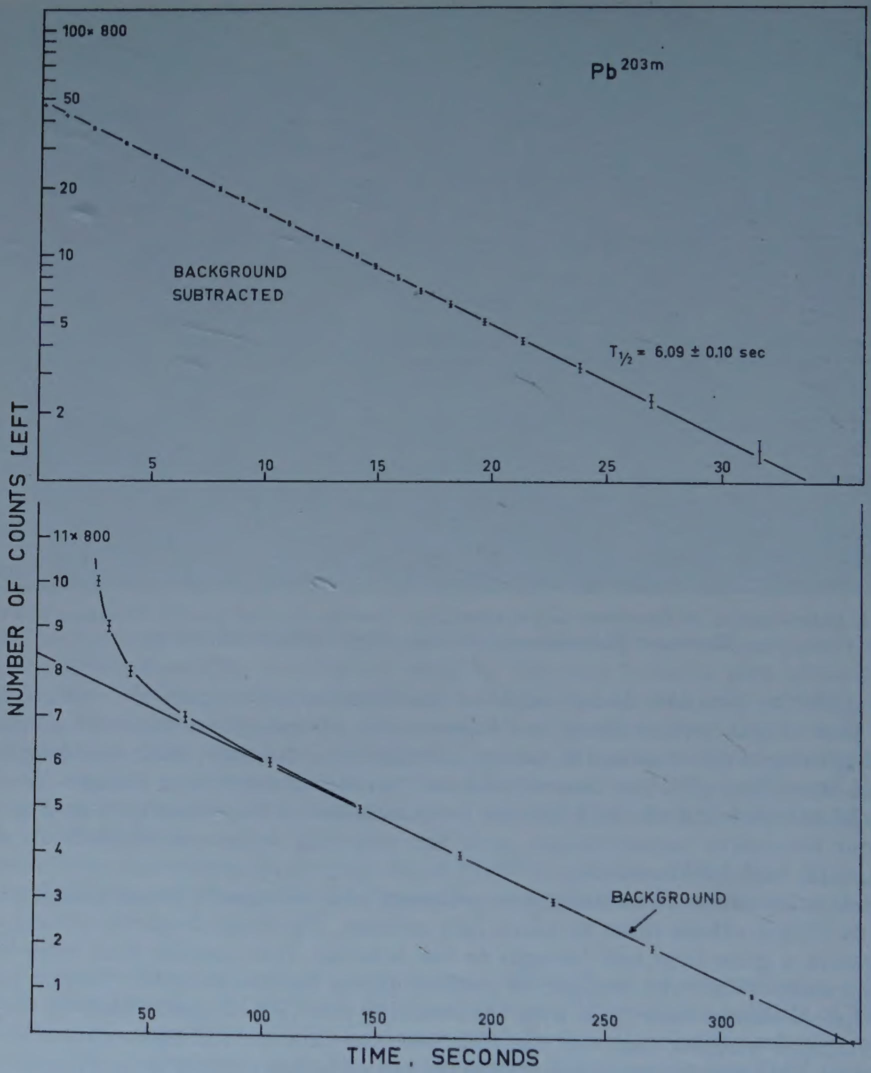

Fig. 5. Decay of Pb^{203m}. Lower curve in linear scale gives background and upper curve in logarithmic scale gives half-life.

530 ± 50 μsec. or 65 ± 5 μsec. The author was also interested in investigating more carefully the possibilities of the "oscilloscope method" for the determination of half-lives of the order of microseconds.

Measurements of delayed coincidences in the actual time region have been made e.g. by Alburger [5] who used an oscilloscope and investigated the time distribution of pulses along the trace by means of a photomultiplier. In this way he found the half-life of the 7 − state in Pb^{206} to be 145 ± 15 μsec. Tove [11] has recently measured the same half-life by means of an electronic time analyzer and found 123 ± 4 μsec.

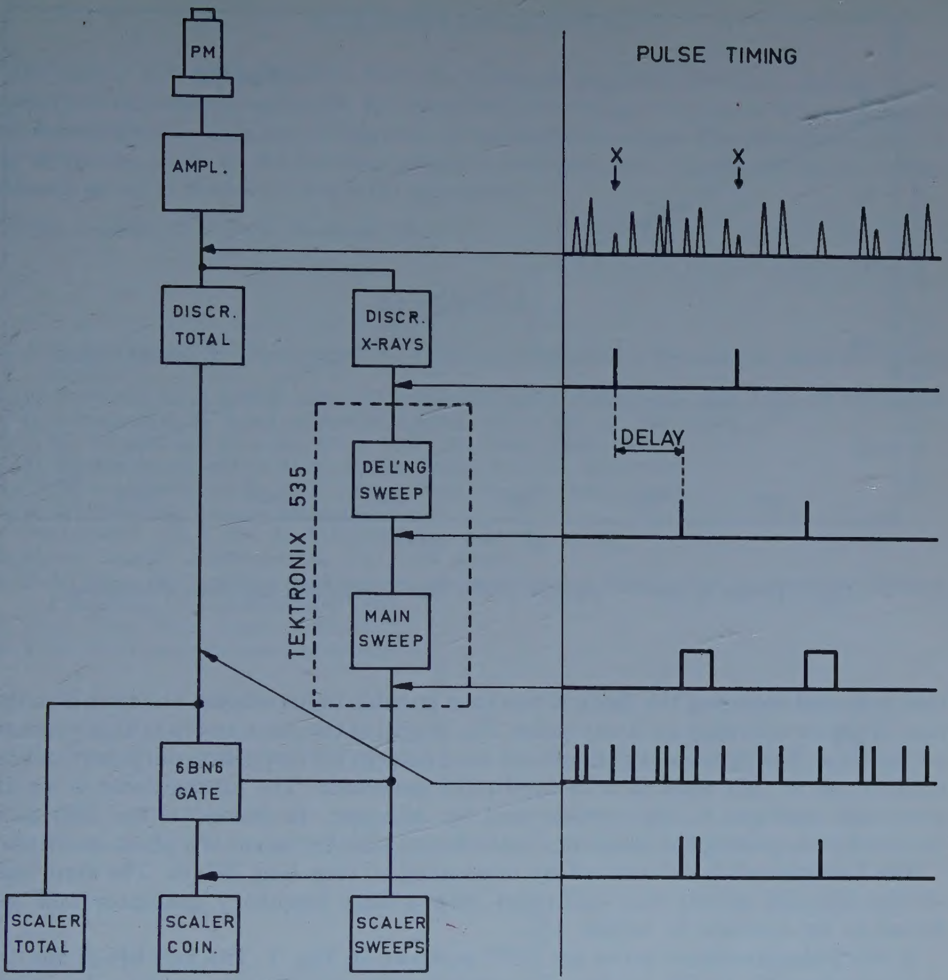

Fig. 6. Block diagram of apparatus used for delayed-coincidence measurements. Time relations are indicated in the right part of the figure.

The present measurements were made in the following way (Fig. 6). A scintillation detector with the sample placed in a hollow crystal ($\varnothing\ 1\frac{3}{4}'' \times 2''$, hole diam. 5 mm) was used. This type of crystal was preferred because it offers the high solid angle which is desirable in coincidence work. Two single-channel discriminators inspected the pulses from the detector. One discriminator passed all pulses including the X-ray pulses to a gate consisting of a 6BN6 tube. The second discriminator selected only the X-ray pulses and triggered the pulse-delaying circuit of a Tektronix oscilloscope type 535. The delayed pulse from that circuit triggered the main sweep of the same oscilloscope. A positive gate pulse of essentially the same length as the main sweep is available and was used to open the 6BN6 gate. By counting the pulses passing

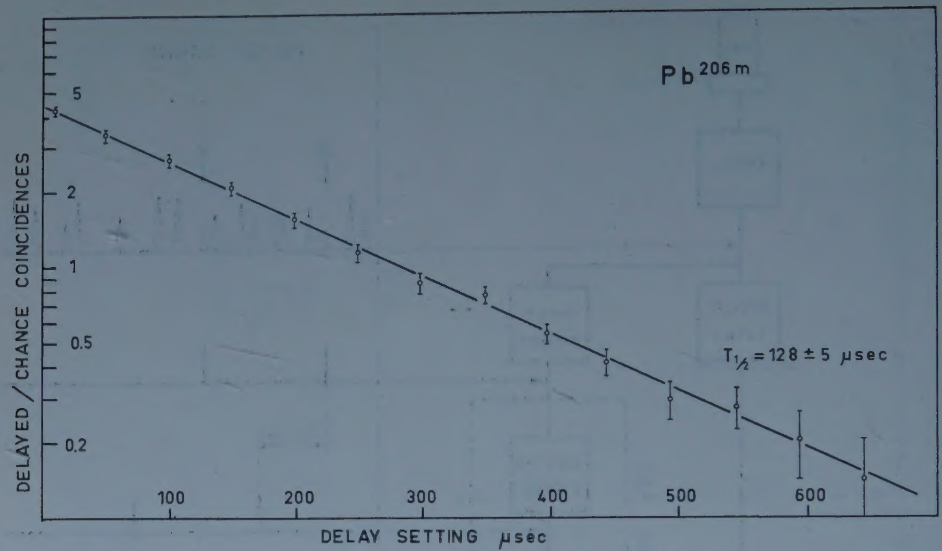

Fig. 7. Decay of the 7 – state in Pb^{206}. The background has been subtracted.

that gate and changing the delay it was thus possible to investigate the time distribution of pulses following an X-ray pulse. The length of the main sweep is kept constant all the time. The cathode ray tube is not used (except for inspection purposes) and the oscilloscope is only used as a delayed-pulse generator. The arrangement is an all-electronic analogue to the system used by Alburger. It eliminates the difficulties involved in inspecting the screen of a cathode-ray tube by means of a photo-multiplier.

The background is determined by measuring at very long delays. The time scale of the delaying circuit was calibrated with a high frequency generator and was found to be accurate to within 1%.

A delayed-coincidence curve for Pb^{206} is shown in Fig. 7. The half-life of the 7 – state was found to be 128 ± 5 μsec. The error is estimated from the statistical accuracy of the points. As can be seen from the curve, the ratio between delayed and chance coincidences was about 4 at small delays.

The result of this investigation shows that the measurement of delayed coincidences in the microsecond region can easily be made by means of standard electronic equipment, available in most laboratories.

A similar measurement was made with a Pb^{202} sample to determine the half-life of the 7 + state in Tl^{202}. As can be seen from the decay scheme, Fig. 1 A, only 10% of the disintegrations go to Tl^{202}. The number of chance coincidences therefore becomes high compared with the number of events associated with the 7 + level. With the best energy selection that could be found, the ratio between delayed and chance coincidences was about one, and the half-life determined in this measurement was 600 ± 200 μsec. Quite recently Tove [12] measured this half-life and found the value 475 ± 20 μsec. It is thus evident that the half-life 530 ± 50 μsec found by Vegors and Axel is associated with the 7 + level in Tl^{202}.

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